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Enhancement of T_c due to pressure application in $LaFeAsO_{1-x}H_x$ studied by NMR

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Abstract
The prototypical iron-based high-T_c pnictide $LaFeAsO_{1-x}H_x$ has superconducting double domes in the electronic phase diagram. Pressure application merges the double domes into a single dome, and the minimum T_c of 26 K observed at x=0.2 goes up to 45 K by applying a pressure of 3.0 GPa. ^{75}As nuclear magnetic resonance was performed at 3.0 GPa to investigate the high-T_c mechanism. The relaxation rate divided by temperature, $1/T_1T$ exhibits a plateau just above T_c and the value is enhanced by applying pressure. The plateau indicates that $1/T_1$ can be described by the Korringa relation, which suggests that a key factor to raise T_c is the density of states. So far as our results are concerned, spin fluctuations are not essential to achieve high T_c over 45 K.

Keywords: FeAs superconductors, NMR, high pressure, spin fluctuation

1 Introduction

The prototypical iron-based high-T_c pnictide $LaFeAsO_{1-x}H_x$ ($0 \leq x \leq 0.6$) exhibits a novel electronic phase diagram: superconducting (SC) double domes emerge upon electron (H) doping following the stripe-type antiferromagnetic (AF) ordering [1]. The AF phase and the first SC dome in a lightly H-doped regime are common to other iron-based systems. The second AF ordering [2] emerges upon further electron doping following the second SC dome, which is unique to this system and owes the capability of electron doping. Intriguingly, pressure application merges the double domes into a single dome, and the minimum T_c of 26 K observed at x=0.2 goes up to 45 K by applying a pressure of 3.0 GPa [1]. Pressure application is equivalent to rare-earth replacement [3]. In fact, the T_c value at 3.0 GPa is almost the same with that for the Ce1111 series, and is close to the highest $T_c(\approx 55K)$ which has been recorded in the

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Sm1111 series [4, 5]. The R1111 (R=Ce, Pr, Nd and Sm) series has higher T_c than the La1111 series, however, rare-earth elements other than La are magnetic, and thus only FeAs layers of the La1111 series are free from magnetic fluctuations originating from rare-earth elements. Therefore, the La1111 series under high pressure offers a good opportunity to determine the high-T_c mechanism. The superconducting mechanism via AF spin fluctuations has been often pointed out as a major candidate of the high-T_c mechanism especially for the 122 series. A primary concern is whether the T_c enhancement in the La1111 series has some relationship to AF spin fluctuations. To address this problem, we investigated low-energy AF spin fluctuations via nuclear magnetic resonance (NMR) measurements.

2 Experimental conditions

The samples were synthesized using a high-pressure apparatus. The H concentration was verified using thermal desorption spectroscopy (TDS), and was almost the same with the nominal concentration. We performed NMR measurements using a conventional coherent-pulsed NMR spectrometer and a 8-Tesla magnet. The relaxation time T_1 was measured by the conventional saturation-recovery method. Pressure was applied by using a NiCrAl-CuBe hybrid piston-cylinder-type pressure cell and was calibrated by ruby fluorescence measurements [6]. Ruby powders were glued on the top of a fiber of 0.25mm outer diameter, and the fiber was inserted in the sample space with the NMR coil, which makes in-situ pressure calibration possible. The length of the NiCrAl cylinder is 30 mm, and the inner and outer diameters are 6 and 16 mm, respectively. The cylinder is covered by a CuBe sleeve with an outer diameter of 40mm. The pressure limit applicable to this cell exceeds 4.0 GPa. We used Fluorinert FC-70 and FC-77 as a pressure mediation liquid. The mixture remains hydrostatic up to 10 GPa. We measured T_c from frequency detuning of the NMR tank circuit, and the relaxation rate divided by temperature (T) $1/T_1T$.

3 Experimental results

3.1 NMR spectra

The field-swept NMR spectra were measured at a frequency (ν_0) of 45.1 MHz. Figure 1 shows the signals corresponding to the central transition of $I = 3/2$ ($I = -1/2 \Leftrightarrow 1/2$) measured at 3.0 GPa and at ambient pressure. The double-peaks structure originates from the second order nuclear quadrupole interaction, and the lower-field peak comes from the powder samples whose iron-basal planes are parallel to the applied field. The spectral intensity at the field h measured from the free resonant field H_0 is given as

$$I(h) \propto \int\int \delta(h - (2\pi/\gamma_N)\nu(\theta))d(cos\theta)d\theta$$

where θ is the angle between h and the c axis, which is perpendicular to FeAs planes, and γ_N is the gyromagnetic ratio of ^{75}As (7.292MHz/T). In the case that anistropy of the electric field gradient is zero ($\eta = 0$), the frequency shift $\nu(\theta)$ from ν_0 is given as

$$\nu(\theta) = -\frac{3\nu_Q^2}{16\nu_0} sin^2\theta(9cos^2\theta - 1).$$

In fact, η is nonzero ($\eta \approx 0.1$), and the formula of $\nu(\theta)$ is more complicated compared to the η=0 case. As seen from Fig. 1, the pressure application hardly causes prominent changes in

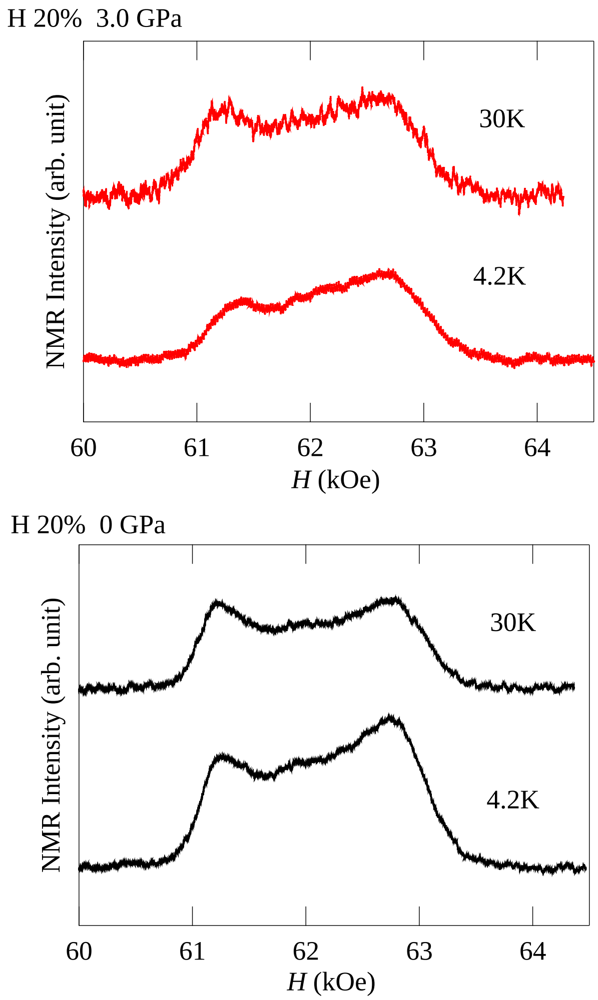

Figure 1: ^{75}As signals corresponding to the central transition ($I = -1/2 \Leftrightarrow 1/2$) measured at 3.0 GPa and at ambient pressure. The relaxation time T_1 was measured at the lower-field peak. The signals at this peak come from powders whose iron-basal planes are parallel to the applied field.

the spectral pattern, and thus $\nu(\theta)$ and η are unchanged by applying pressure at this doping level.

3.2 Relaxation rate

Figure 2 shows $1/T_1T$ at 3.0 GPa and at ambient pressure. At ambient pressure, $1/T_1T$ exhibits monotonous increase with increasing temperature except for a plateau observed in a narrow-T range. The onset temperature where $1/T_1T$ deviates from the plateau is regarded as T_c ($\approx$

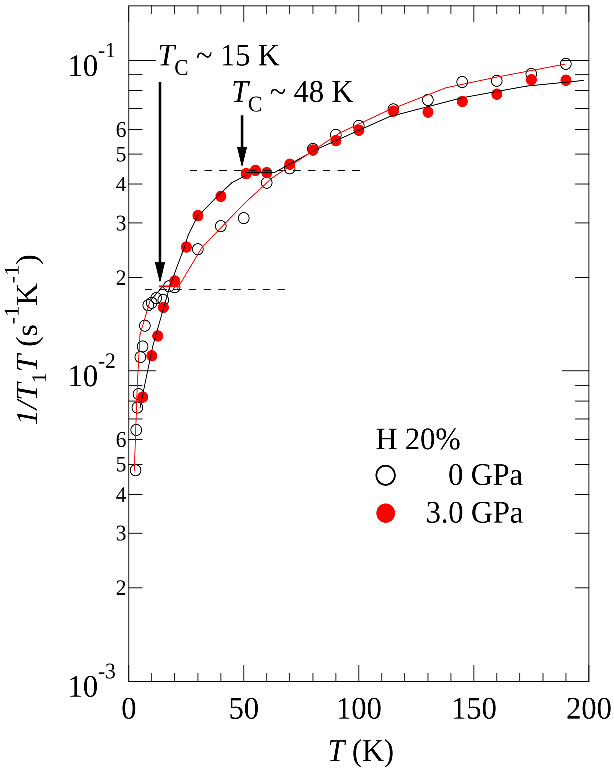

Figure 2: Relaxation rate divided by temperature $1/T_1T$ of ^{75}As. The measurements were performed at the lower-field peaks in Fig. 1.

15 K). The T_c value determined from the frequency detuning is about 20 K. The monotonous increase in $1/T_1T$ above T_c is not due to AF spin fluctuations, but due to the density of states involved only at high temperatures. Such T dependence is not rare and has been also observed in $K_yFe_{2-x}Se_2$ [7]. As seen from Fig. 2, pressure application hardly causes a qualitative change in $1/T_1T$ except for the enhancement of $1/T_1T$ at the plateau. T_c is enhanced from 15 K to 48 K by applying a pressure of 3.0 GPa. At 3.0 GPa, the plateau is observed in a wider T range than at ambient pressure. Figure 3 shows $1/T_1T$ for the 14% F-doped $LaFeAsO_{1-x}F_x$ measured at 3.0 GPa and at ambient pressure [8, 9]. Overall features are the same with those observed for the 20% H-doped samples: a plateau of $1/T_1T$ just above T_c and monotonous T dependence of $1/T_1T$ at high temperatures, and so on. The plateau is seen more clearly compared with the case of the 20% H-doped samples.

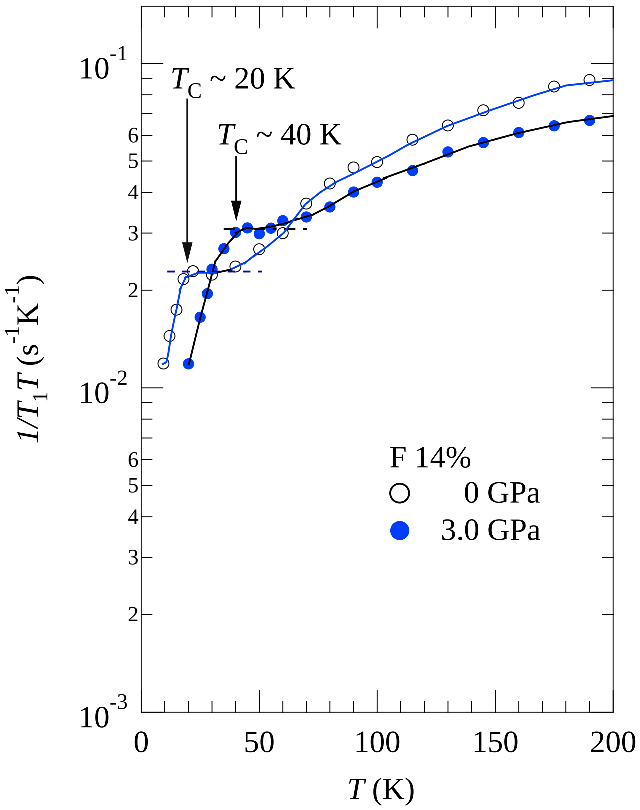

Figure 3: $1/T_1T$ of ^{75}As for the14% F-doped $LaFeAsO_{1-x}F_x$ samples. The measurements were performed at the lower-field peaks in Fig. 1.

4 Discussion

The T_c enhancement is well understood by considering equivalency between pressure application and rare-earth replacement. A pressure of 3.0 GPa corresponds to the Ce1111 series $CeFeAsO_{1-x}F_x$ at ambient pressure. When replacing La with other elements with smaller radius, the FeAs local structure approaches regular tetrahedra, and the lattice constants shrink as well. T_c itself is determined by the local structure of FeAs tetrahedra. The problem is whether AF fluctuations are involved in raising T_c up to $\approx 50K$. So far as our results are concerned, the T_c enhancement is caused without predominant spin fluctuations. Among a variety of FeAs iron-based superconductors, only the 1111 series reaches a high T_c above 50 K so far. For 40% H-doped samples, T_c reaches 48 K at 3.0 GPa that is close to 55K [4, 5], the highest T_c ever known in iron-based superconductors.

One can compare the results of Fig. 2 with those of Fig. 3, because 14% F doping is close to 20% H doping in the electronic phase diagram. As seen from both figures, T_c has a strong

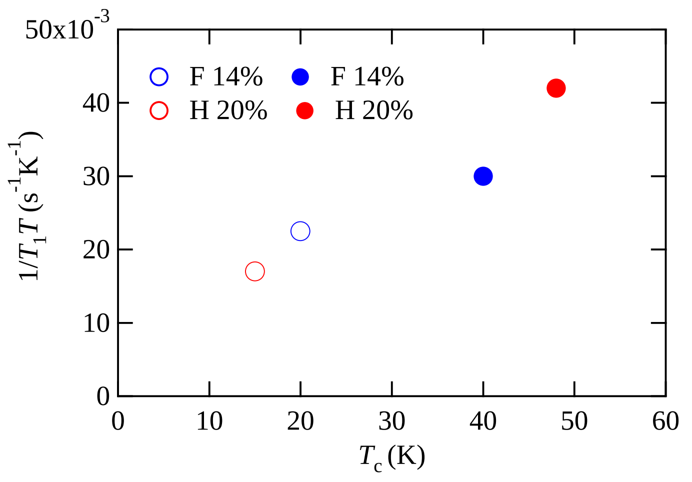

Figure 4: T_c vs. $1/T_1T$ at the plateaus in Figs. 2 and 3.

correlation with $1/T_1T$ at the plateau. In general, $1/T_1T$ is described by the Korringa relation in the case of conventional metals in which strong spin fluctuations are absent. In the Korringa relation, spin correlations are included in the spin Stoner enhancement factor α_s, and $1/T_1T$ is proportional to square of the density of states $N(\varepsilon_F)$ and the hyperfine coupling constant A_{hf}, namely

$$\frac{1}{T_1T} \propto A_{hf}^2 \frac{N(\varepsilon_F)^2}{1-\alpha_s}.$$

In general, α_s gives a measure of spin fluctuations: however, predominant spin fluctuations are absent in the present case, and thus α_s can be regarded as the same between 14% F and 20% H doping. If α_s would be different for the two doping levels, $1/T_1T$ would shift by a factor for a whole temperature range. The fact that $1/T_1T$ is almost the same at high temperatures around 200 K indicates that α_s and A_{hf} are unchanged between the two doping levels. Therefore, a key factor to raise T_c is attributable to the enhancement of $N(\varepsilon_F)$, as shown in Fig. 4.

Superconducting mechanism due to AF fluctuations has been regarded as a candidate to explain high T_c in iron-based superconductors. In this case, spin fluctuations would have a correlation with T_c: however, a high T_c value ($\approx$ 48 K) has been achieved without observable spin fluctuations. This fact would give credence for superconducting mechanism via orbital fluctuations, another candidate for the high-T_c mechanism [10].

5 Conclusion

We performed NMR measurements on $LaFeAsO_{1-x}H_x$ at 3.0 GPa. The pressure application raises T_c from 15 K at ambient pressure to 48 K without an observable enhancement of AF fluctuations. The results of $1/T_1T$ indicate that an enhancement of $N(\varepsilon_F)$ is a key factor to raise T_c and spin fluctuations are unlikely to be involved in the superconducting mechanism.

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